Experimental Report

19/04/2014

Proposal:	**DIR-107**
Council:	10/2012
Title:	Spin Susceptibility of UPt3

This proposal is a new proposal

Researh Area:

Main proposer: **GANNON William**

Experimental Team: RASTOVSKI Catherine
GANNON William
HALPERIN william
ESKILDSEN Morten Ring
HANSEN Ursula Bengaard

Local Contact: STUNAULT Anne

Samples: UPt3

Instrument	Req. Days	All. Days	From	To
D3	7	7	28/05/2013	04/06/2013

Abstract:

Spin Susceptibility of UPt_3

W. J. Gannon and W. P. Halperin
Department of Physics and Astronomy, Northwestern University, Evanston, IL 60660 USA

M. R. Eskildsen
Department of Physics, University of Notre Dame, Notre Dame, IN 46556 USA

Pengcheng Dai
Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996 USA

U. B. Hansen and K. Lefmann
Niels Bohr Institut, University of Copenhagen, Copenhagen, Demnark

A. Stunault
Institut Max von Laue-Paul Langevin, 38042 Grenoble, France
(Dated: April 19, 2014)

This report covers beamtime DIR-107. Polarized neutron diffraction measurements were made on the heavy fermion superconductor UPt_3 using the D3 two-axis diffractometer. The magnetization of UPt_3 was measured along the crystal c-axis at two fields across the transition between the normal state and the superconducting A-, B-, and C- phases. No change in magnetization was detected across these transitions. Field dependence measurements at low temperatures also indicate no decrease in spin susceptibility in the superconducting state from the normal state. These results imply that the Cooper pair spins in UPt_3 are in an equally spin paired triplet state and that the spin quantization axis follows magnetic fields larger than 0.2 T.

With its multiple superconducting phases, UPt_3 has long been a paradigm for unconventional superconductivity, most likely having an f-wave pair orbital state [1–3]. For unconventional superconductors, the temperature dependence of the spin susceptibility is an important signature of the superconducting state as, it contains information about the spin state of the Cooper pair electrons. In a previous experiment on D3 at ILL (proposal TEST-1963), this collaboration found that the spin susceptibility was unchanged across the superconducting-normal transition when measured along the crystal a-axis, implying that the Cooper pairs were in an equally spin paired triplet state [4]. This result was expected, as a wide variety of experimental probes and theory [3] suggest that UPt_3 is indeed a spin-triplet superconductor.

What remains at issue, however, are the details of that triplet state. In this regard, there are two contradictory results: On the one hand, ultrasonic measurements of the upper critical field [5] imply Pauli limiting along the crystal c-axis, suggesting that the spin state is described by an equal spin paired triplet with the equal spin pairs confined to be in the crystal basal plane [1, 6]. On the other hand, the NMR Knight shift measured across T_c [7, 8] is temperature independent for all field directions with no suppression in the superconducting state, implying that the Cooper pair spins are in an equally spin paired triplet and that the pairs are free to rotate with only a small applied field (as small as 0.2 T in those experiments) in any direction [2].

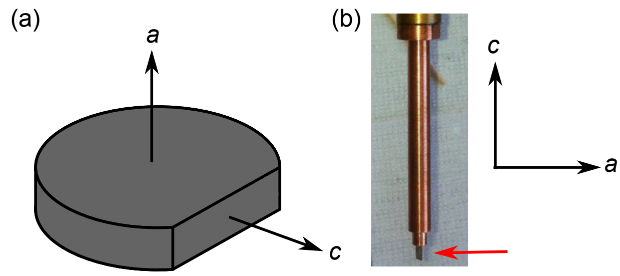

FIG. 1. (a) A schematic of the UPt_3 sample used for this experiment. Crystal axes are shown by black arrows. (b) A picture of the sample, mounted on the cold finger. Sample is indicated by the red arrow.

This conflict was not resolved by our previous experiment [4], which matched the Knight shift results well, but can be interpreted to support both theories, as it did not test the ability of the triplet spins to rotate out of the basal plane. Because of this and the fact that the Knight shift measurements are only sensitive to a London penetration depth from the sample surface – where scattering at the surface can mask bulk behavior [9] – it is necessary to measure the magnetization of UPt_3 along the crystal c-axis with a bulk probe such as neutron scattering to test wheather or not the Cooper pair spins are free to rotate.

Our UPt_3 sample was a high quality single crystal (RRR $\approx$ 900) of 0.4 g total mass. The crystal was attached with silver epoxy to a copper cold finger,

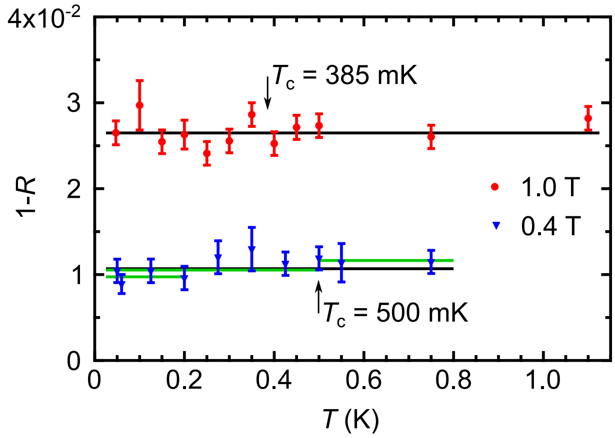

FIG. 2. The flipping ratio R as a function of temperature for the (100) nuclear Bragg reflection at two fields: 0.4 T (blue triangles), and 1.0 T (red circles). Black lines show the average value of 1-R at each field. Green lines indicate the average of 1-R for the 0.4 T data for the temperature range over which the line is drawn.

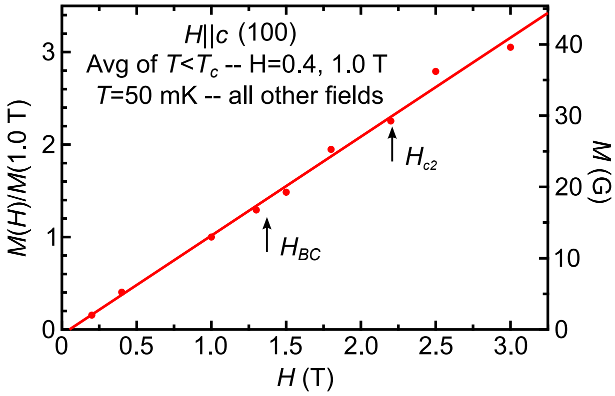

FIG. 3. The field dependence of the magnetization with fields along the c-axis measured by neutrons. The right axis shows M in absolute units, while the left axis shows M normalized to M measured at 1.0 T. The average values of the magnetization for $T < T_c$ were used here for $H = 0.4$ and 1.0 T. To within the accuracy of our data, $\chi = M/B$. Consistently, the zero field intercept of a linear fit to our data is $M(H)/M(1.0T)$ intercept is -0.05. Field of the A-B phase transition and H_{c2} are indicated by black arrows.

aligned in the $a^* - b^*$ scattering plane. The cold finger was mounted to the mixing chamber of a dilution refrigerator, which was cooled inside of a vertical superconducting magnet on the D3 two-axis diffractometer at ILL. The detector was positioned at a nuclear Bragg reflection and neutrons of wavelength 0.825 Å were incident on the sample.

This experiment measures the flipping ratio R, defined as the ratio of scattering cross sections for neutrons with spins parallel and anti-parallel to an applied magnetic field with arbitrary final spin state [10]. When our experimental geometry is taken into account, the flipping ratio reduces to:

$$1 - R = \frac{2\gamma r_0}{\mu_B} \frac{M_{||}(\vec{\kappa})}{F_N(\vec{\kappa})}. \qquad (1)$$

where γr_0 is the neutron gyromagnetic ratio multiplied by the classical radius of the electron, μ_B is the Bohr magneton, and $F_N(\vec{\kappa})$ is the nuclear structure factor of the Bragg reflection being measured. $M_{||}(\vec{\kappa})$ is the component parallel to the applied magnetic field of the Fourier transform of the real space magnetization $M(\vec{r})$. Since the magnetic field is always along the crystal c-axis in this experiment, we are always measuring the magnetization in this direction, perpendicular to the basal plane.

Fig. 2 shows 1-R measured at the (100) nuclear Bragg reflection for two different applied fields. These fields were chosen such that they each cut through a different portion of the superconducting phase diagram. The 0.4 T data enter the superconducting state in the A-phase at $T_c = 500$ mK, then enter the superconducting B-phase at a temperature of 425 mK. The 1.0 T data enter the superconducting state at $T_c = 385$ mK, very near the

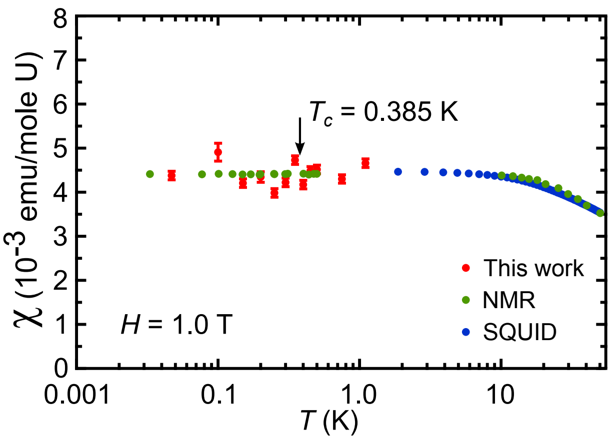

FIG. 4. Magnetic susceptibility measured by polarized neutron diffraction measured in a 1.0 T field (red circles) multiplied by a constant factor of 2.35, plotted with the susceptibility measured along the c-axis in the normal state with SQUID (blue circles), and the Knight shift from from Refs. 7 and 8 (green circles).

A-B-C tetracritical point and are always in the B-phase below T_c. Measurements along the c-axis have lower signal-to-noise ratio than our previous measurements with field along the a-axis [4], as the magnetic susceptibility is a factor of two smaller along the c-axis, reducing the signal size in the current experiment by a factor of four. The temperature dependence of $1 - R$ at the (100) reflection appears to be temperature independent for both fields measured. It is possible that there is a slight decrease in $1 - R$ below T_c in the 0.4 T data. However, it is not possible to determine this definitively, given the size of our error bars. A piecewise fit of the temperature dependence above T_c, below T_c, and at the lowest temperatures (< 200 mK) places an upper limit on the magnitude of the decrease at 16%.

Fig. 3 shows the field dependence of the magnetization measured in this experiment across the entire superconducting phase diagram at low temperatures. The right axis shows shows our measurements in absolute units, while the left axis shows our measurements normalized to the value of the magnetization at 1.0 T. The linearity of the data, including data measured in the normal state above H_{c2} indicates that there is no deviation from $\chi = M/B$. This is consistent with the measurements of the temperature dependence in Fig. 2 and supports the notion that the equal spin pairs are able to rotate out of the basal plane with only a relatively small field (0.2 T).

Fig. 4 shows the superconducting and normal state susceptibility from polarized neutrons measured at the (100) reflection in a 1.0 T field plotted with normal state measurements from a SQUID made on a piece of UPt_3 of similar quality as the sample used in the neutron scattering experiment, as well as the susceptibility calculated from the Knight shift measurements at 1.0 T of Tou *et. al.* [7, 8]. Our neutron data match well with both when the neutron data are multiplied by a factor of 2.35. This discrepancy is attributed to non-zero beam depolarization and the fact that we are measuring M at a non-zero q. The good agreement we find gives confidence that the Knight shift results are accurately representative of bulk behavior.

The temperature independence of these results in the superconducting state indicate that the electron spins in the Cooper pairs in UPt_3 are in an equal spin paired triplet state. Combined with our previous measurements [4], these results would suggest that UPt_3 is a triplet superconductor where the field scale to rotate the pair spins out of the basal plane is rather small, < 0.2 T. This is surprising and somewhat unrealistic, as the most prominent theory to describe this physics [2] requires weak spin-orbit coupling, which is unlikely, owing to the heavy masses of the constituent elements in UPt_3. This theory is also contradicted not only by the Pauli limiting of H_{c2} [5] mentioned earlier, but also by phase sensitive tunneling measurements [11, 12]. That being said, our results indicate that the Knight shift measurements can not be discounted, and a theoretical puzzle remains to account for the spin dependence of the order parameter of UPt_3.

Research support for W. J. G., W. P. H., M. R. E., and P. D. was provided by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Awards No. DE-FG02-10ER46783 (University of Notre Dame and Northwestern University), No. DEFG02-05ER46248 (Northwestern University), and No. DE-FG02-05ER46202 (University of Tennessee). Support for U. B. H. and K. L. was provided by the Danish Research council FNU through the grants DANSCATT and Spin Architechture and Magnetism of single-molecules and extended systems.

[1] J. A. Sauls, Advances in Physics **43**, 113 (1994).
[2] Y. Tsutsumi, K. Machida, T. Ohmi, and M. Ozaki, Journal of the Physical Society of Japan **81**, 074717 (2012).
[3] R. Joynt and L. Taillefer, Reviews of Modern Physics **74**, 235 (2002).
[4] W. J. Gannon, W. P. Halperin, C. Rastovski, M. R. Eskildsen, P. Dai, and A. Stunault, Physical Review B **86**, 104510 (2012).
[5] B. S. Shivaram, T. F. Rosenbaum, and D. G. Hinks, Physical Review Letters **57**, 1259 (1986).
[6] M. J. Graf, S. K. Yip, and J. A. Sauls, Physical Review B **62**, 14393 (2000).
[7] H. Tou, Y. Kitaoka, K. Asayama, N. Kimura, Y. Onuki, E. Yamamoto, and K. Maezawa, Physical Review Letters **77**, 1374 (1996).
[8] H. Tou, Y. Kitaoka, K. Ishida, K. Asayama, N. Kimura, Y. Onuki, E. Yamamoto, Y. Haga, and K. Maezawa, Physical Review Letters **80**, 3129 (1998).
[9] A. A. Abrikosov and L. P. Gor'Kov, Zhurnal Eksperimentalnoi i Teoreticheskoi Fiziki **39**, 480 (1960).
[10] G. L. Squires, *Introduction to the Theory of Thermal Neutron Scattering*, 2012th ed. (Cambridge University Press, England, 1978).
[11] J. D. Strand, D. J. V. Harlingen, J. B. Kycia, and W. P. Halperin, Physical Review Letters **103**, 197002 (2009).
[12] J. D. Strand, D. J. Bahr, D. J. V. Harlingen, J. P. Davis, W. J. Gannon, and W. P. Halperin, Science **328**, 1368 (2010).